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Application of synchrotron X-ray scattering to the understanding of the crystallization of complex mixtures of triglycerides, for a rational design of tailored confectionary products

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Chocolate is a popular product and its supply is increasing, with sales in the UK estimated to be £6.7 billion in 2020, a £200 million increase from 2019. In recent years, confectionary companies have put increasing effort in developing novel recipes to improve the nutritional profile of chocolate and to counteract the increasing price of cocoa butter and address sustainability issues related to some chocolate ingredients. One of this these strategies is the use of cocoa butter equivalents (CBE), which are mixtures of triglycerides from multiple sources (e.g., sunflower oil, mango kernel, sal) that resemble cocoa butter in both physical and chemical properties. Despite being widely used, the crystallization behaviour of many CBEs is still poorly understood. The aim of this work was to develop a fundamental understanding, at the molecular level, of the crystallization behaviour of selected CBEs, and compare it with that of cocoa butter. In order to do so, chromatography was used to determine the composition of CBEs, in terms of fatty acids and triglycerides while the thermodynamic and kinetics of crystallization were studied using polarized microscopy, differential calorimetry and several, unique synchrotron X-ray scattering setups (the multi-capillary holder at Diamond Light Source in the United Kingdom, the combined DSC/SAXS at Elettra Sincrotrone Trieste in Italy and the combined SAXS/rheometry at ESRF in France). The combination of these techniques enabled the determination of crystal properties that affect the sensorial perception of chocolate: namely the type of crystals formed (e.g., polymorphism), their thermal stability and their size and shape distributions. Furthermore, the kinetics of crystallization as a function of CBE composition and the effect of shear were evaluated. The presented multi-technique investigation is the key for a rational design of new chocolate recipes and manufacturing processes.